

# **Ultrafast Processes in Solids Probed by Femtosecond X-ray Pulses**

**Klaus Sokolowski-Tinten**

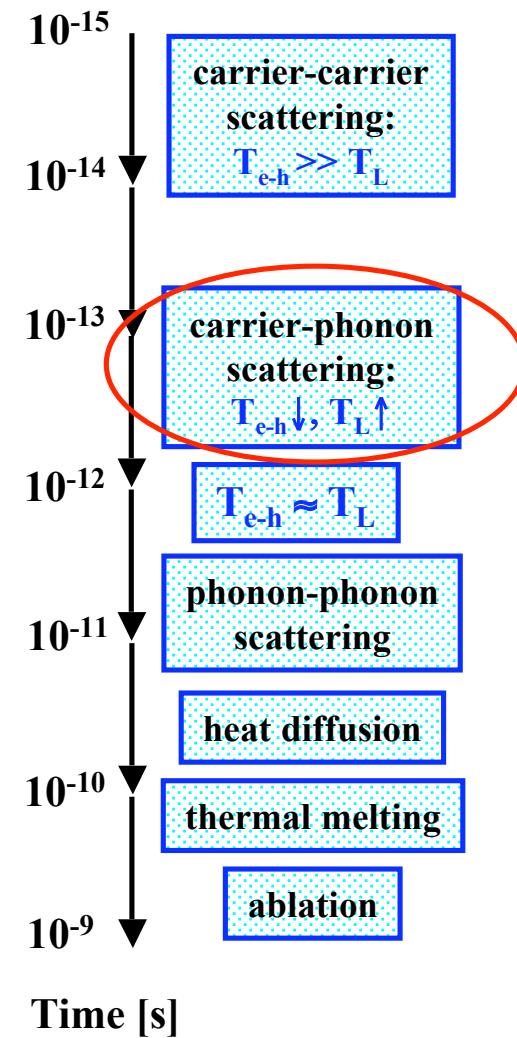
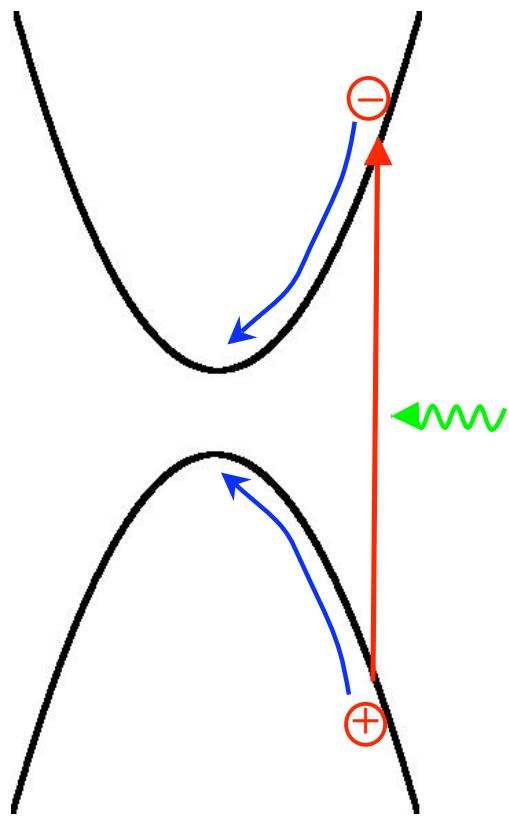
Institut für Experimentelle Physik



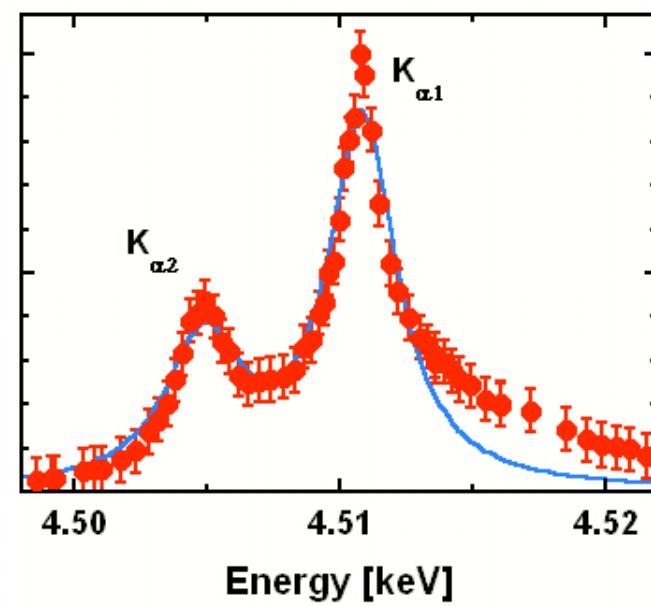
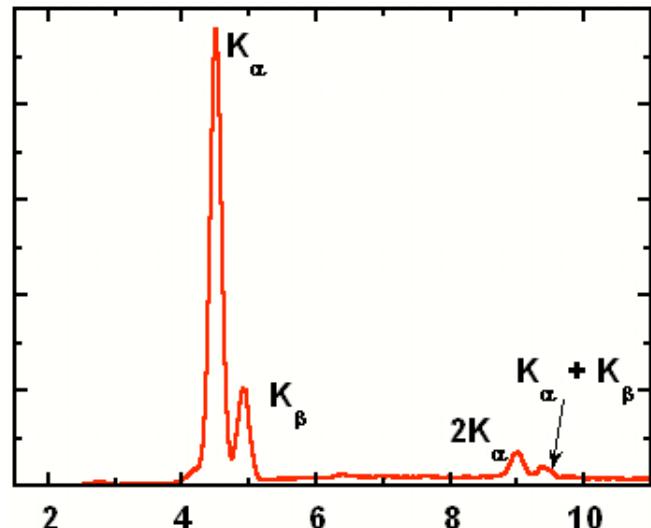
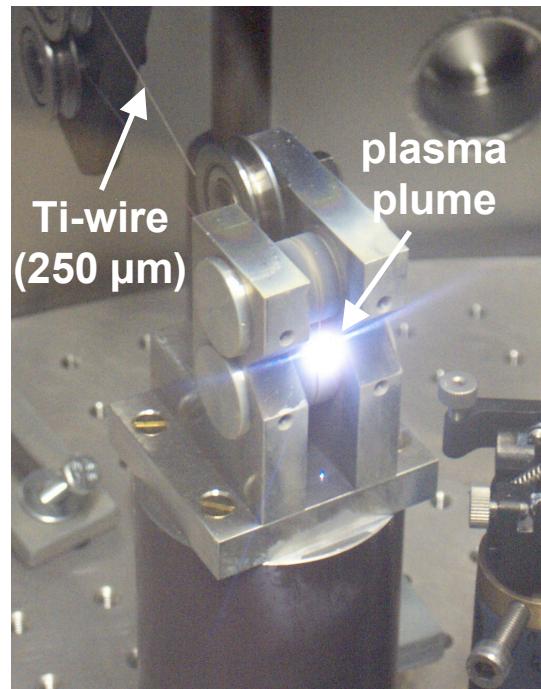
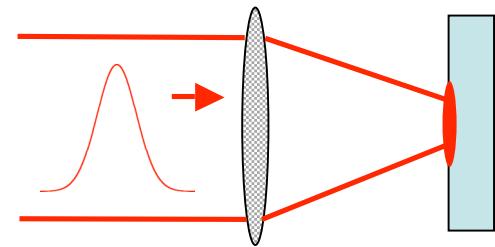
# Outline

- Motivation
- X-rays from laser-produced plasmas
- Ultrafast melting
- Energy relaxation in Germanium
- Coherent optical phonons in Bismuth

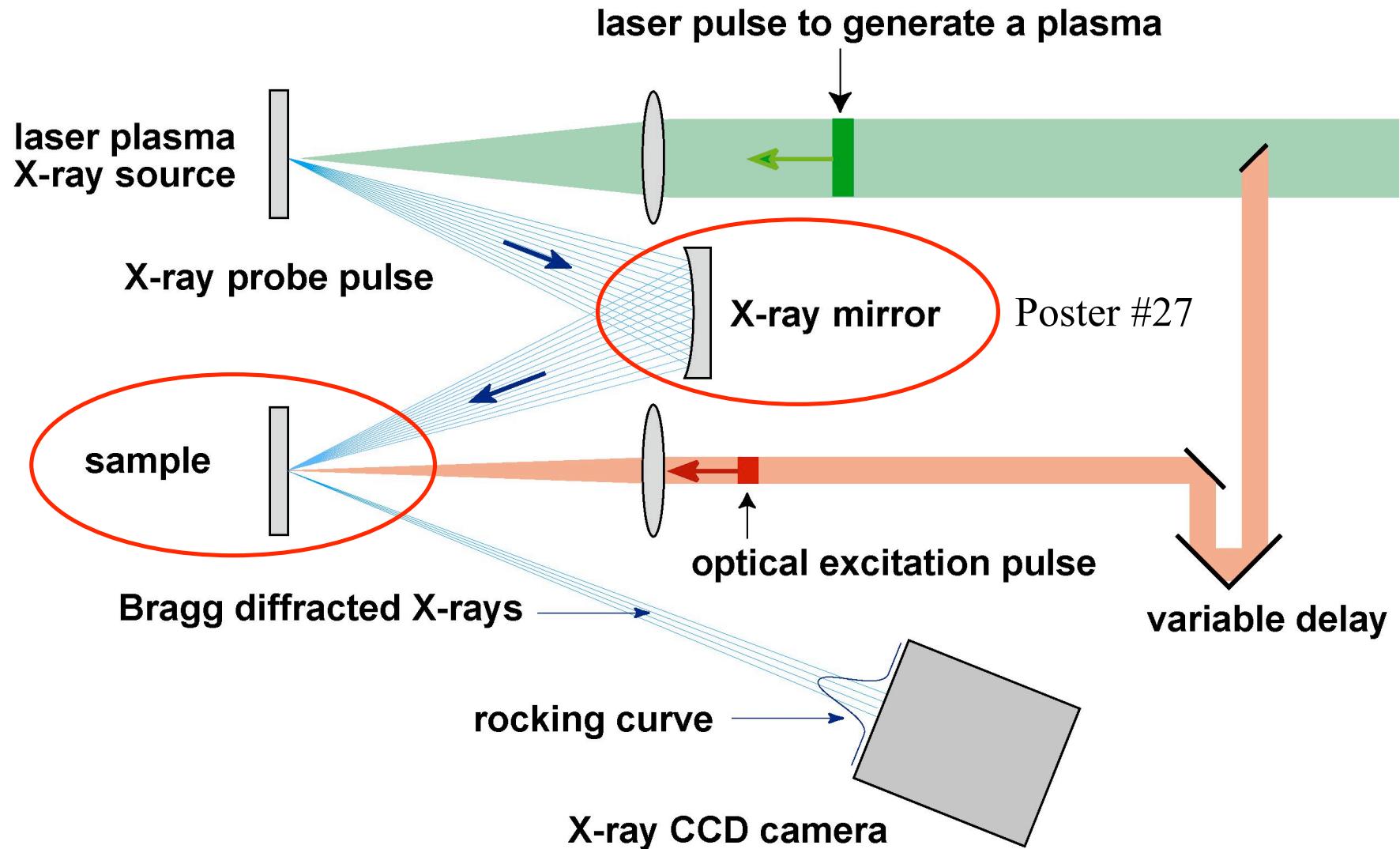
# Relaxation in Optically excited Solids

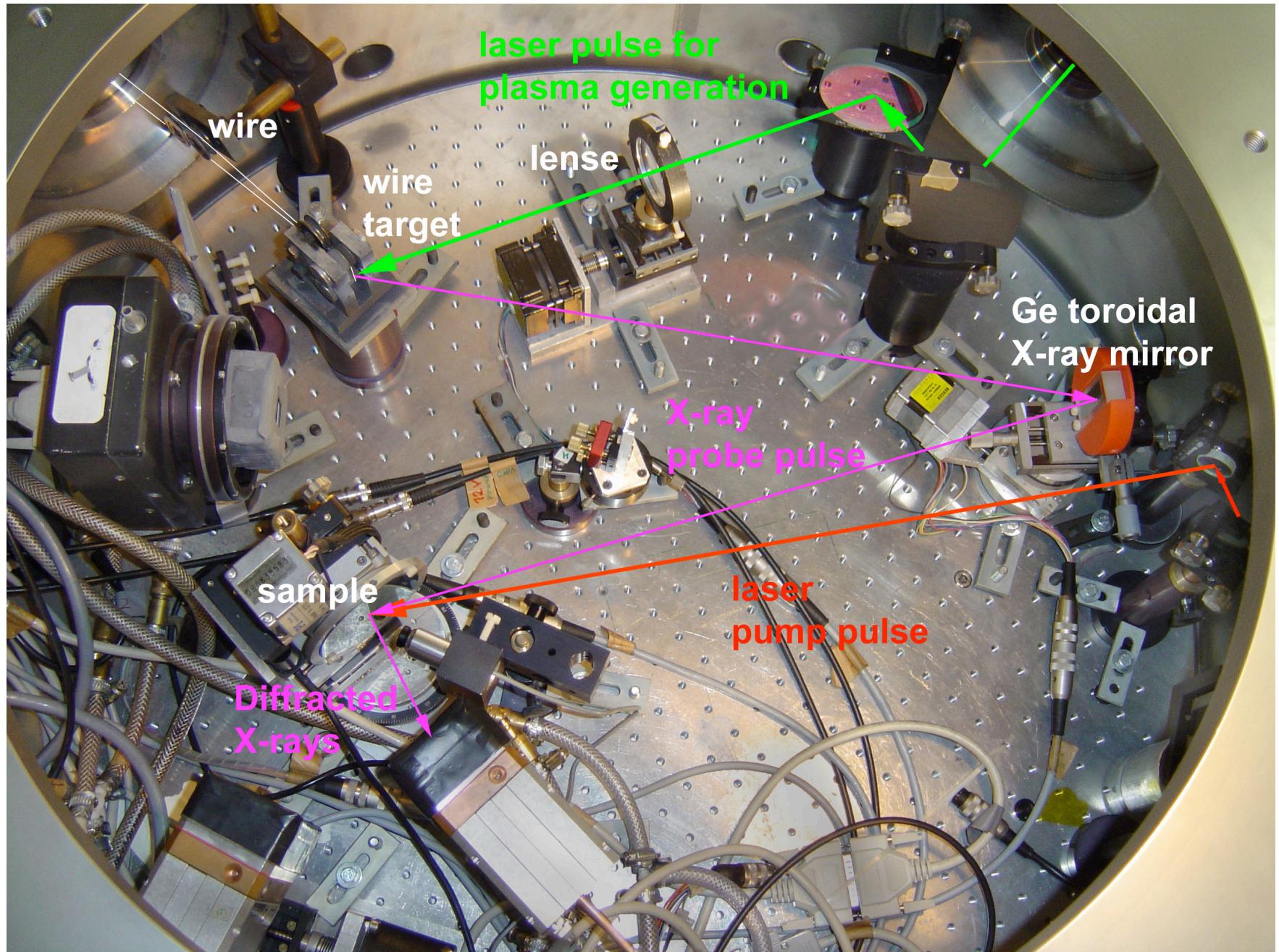


# The X-ray Candle: *Laser-Produced Plasmas*

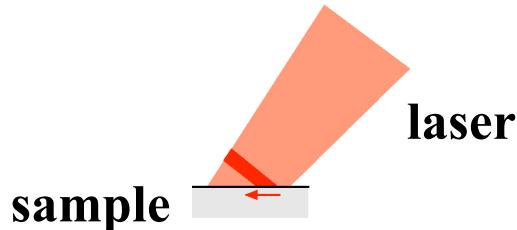


# Ultrafast Time-Resolved X-ray Diffraction (UXRD)





# Ultrafast Melting: Optical Spectroscopy



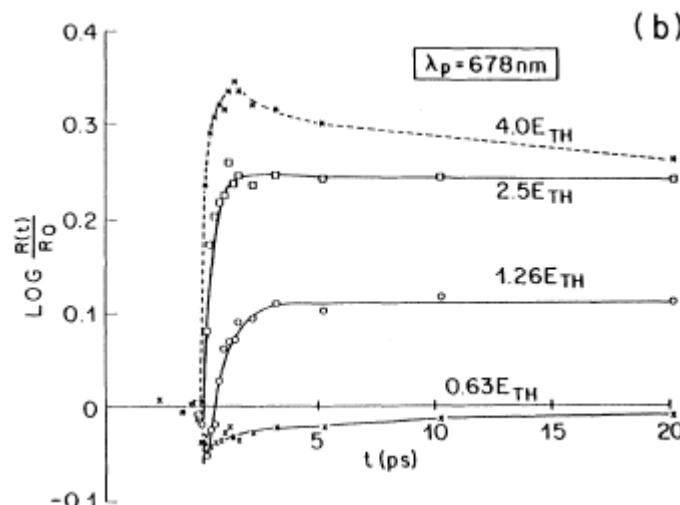
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PHYSICAL REVIEW LETTERS

7 FEBRUARY 1983

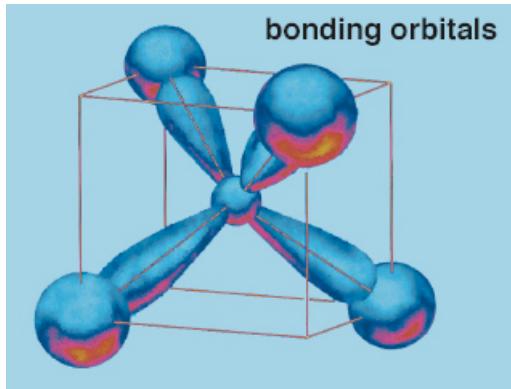
## Time-Resolved Reflectivity Measurements of Femtosecond-Optical-Pulse-Induced Phase Transitions in Silicon

C. V. Shank, R. Yen, and C. Hirlimann

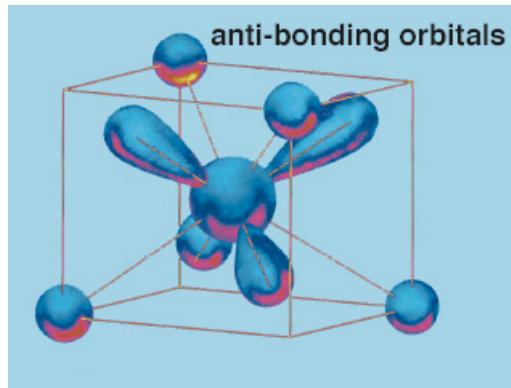
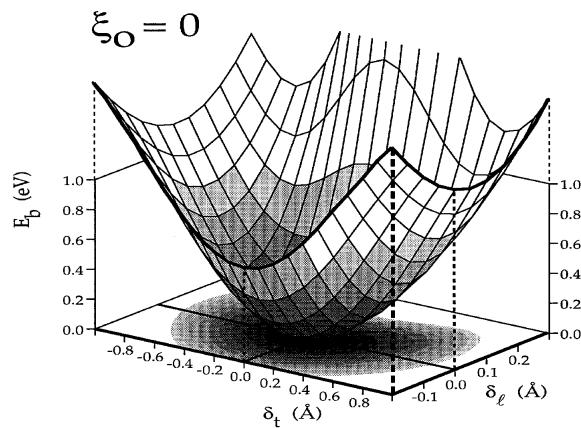


# Non-Thermal Melting of Semiconductors

*chemical:*



*solid state:*



*lattice destabilization*

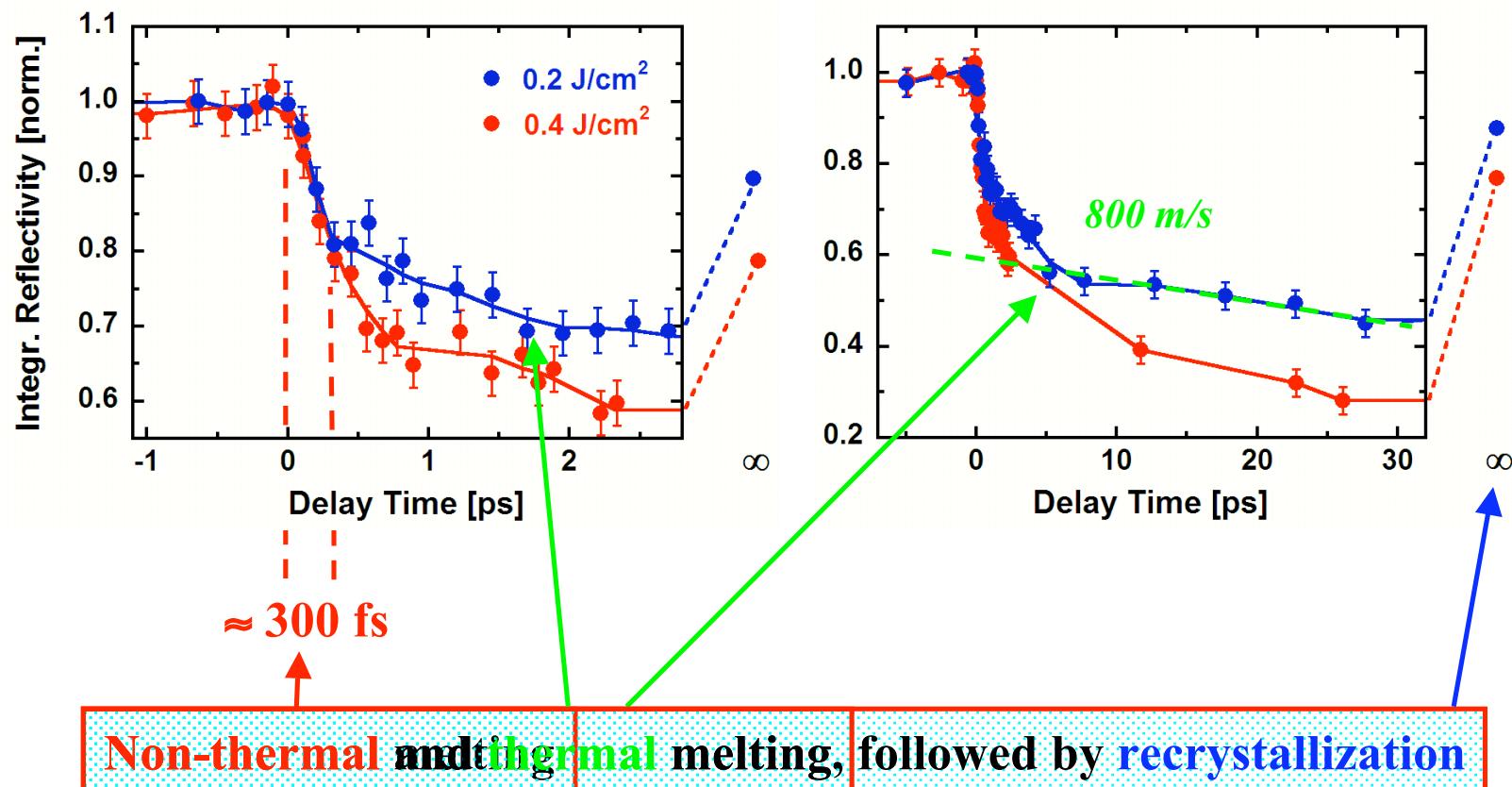
# Non-Thermal Melting: *Test-Case* for TXRD

- Chin et al., Phys. Rev. Lett. **83**, 336 (1999)
- Siders et al., Science **286**, 1340 (1999)
- Lindenberg et al., Phys. Rev. Lett. **84**, 111 (2000)
- Rousse et al., Nature **410**, 65 (2001)
- Sokolowski-Tinten et al., Phys. Rev. Lett. **87**, 225701 (2001)
- Feurer et al., Phys. Rev. E **65**, 16412 (2002)
- Lindenberg, Larsson, Gaffney, Blome, Synnergren, Sheppard, Calemann, Sokolowski-Tinten & SPPS Collab., (2004).

# X-ray Diffraction: Melting of Germanium

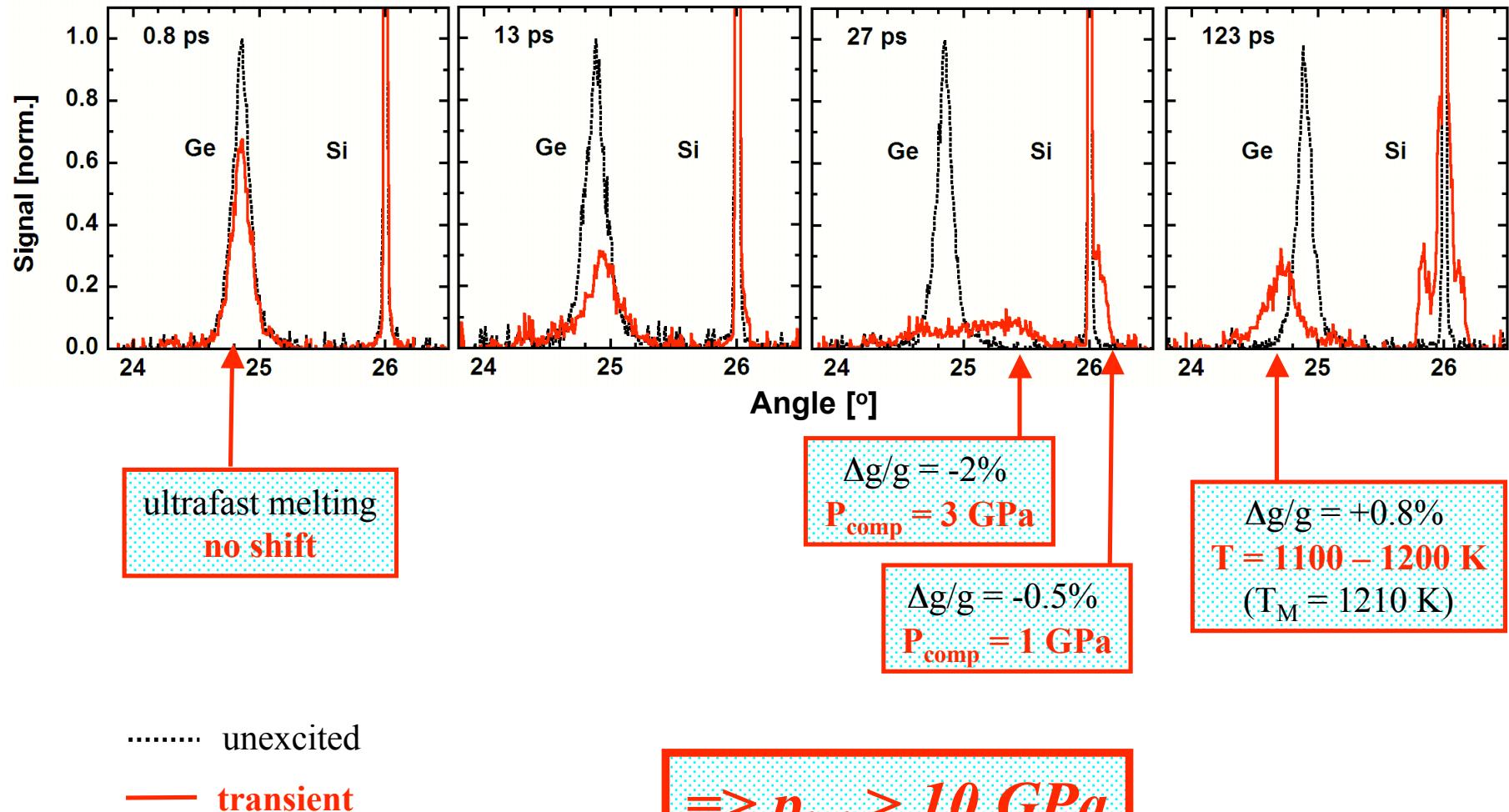
170 nm Ge on Si; (111)-reflection

K.Sokolowski-Tinten et al., PRL **87**, 225701 (2001)

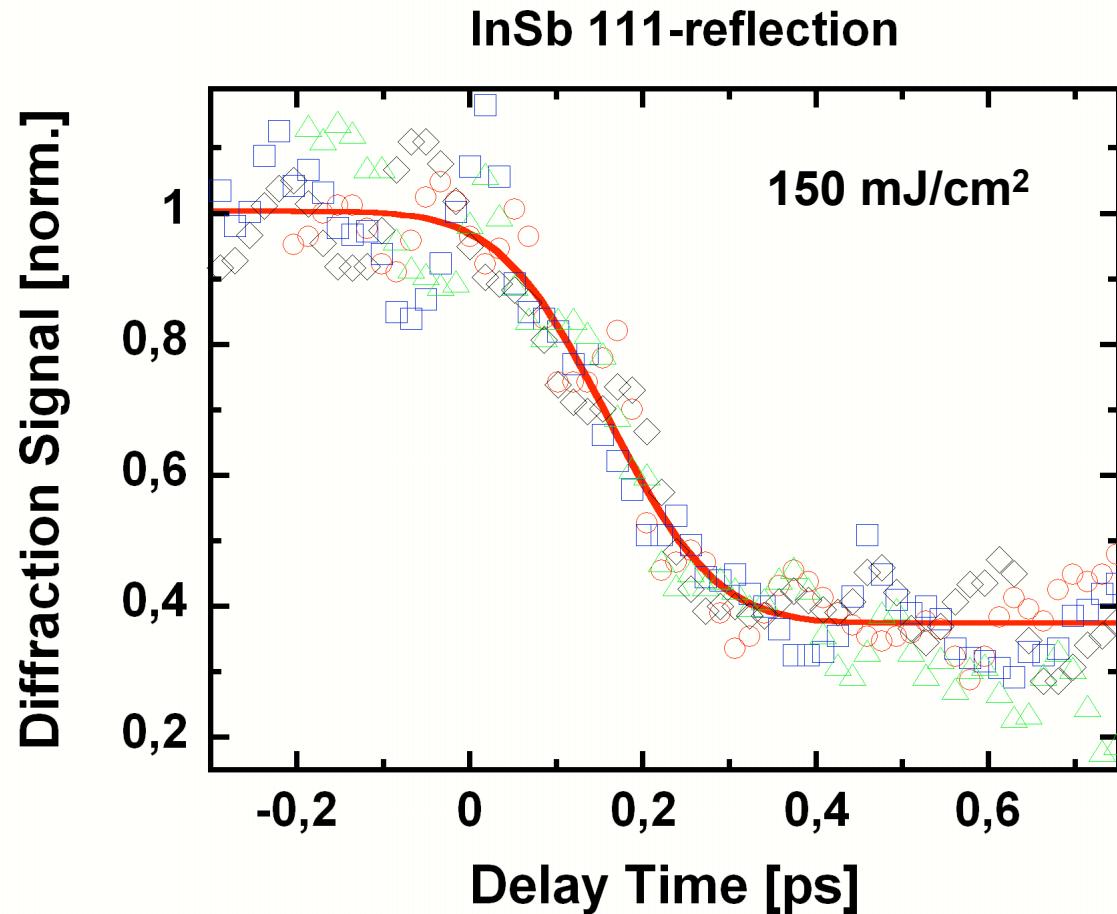


# Diffraction Profiles

170 nm Ge on Si;  $F = 0.2 \text{ J/cm}^2$

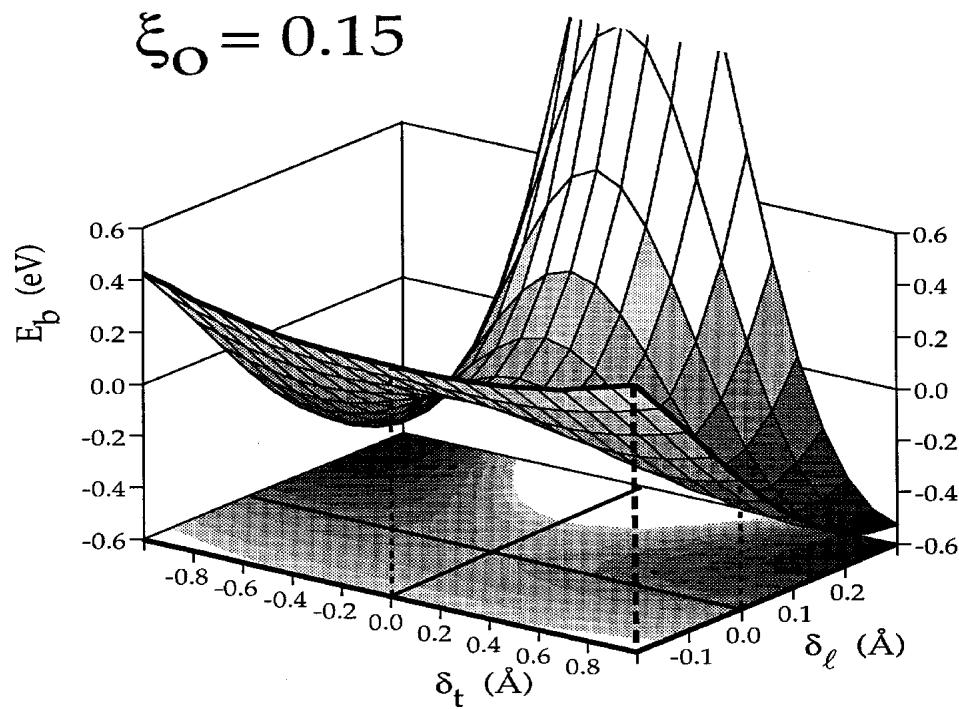


# New Results from SPPS: *Disordering of InSb*



*! first femtosecond x-ray diffraction imaging using an advanced laser-based source !*

# What is next ??

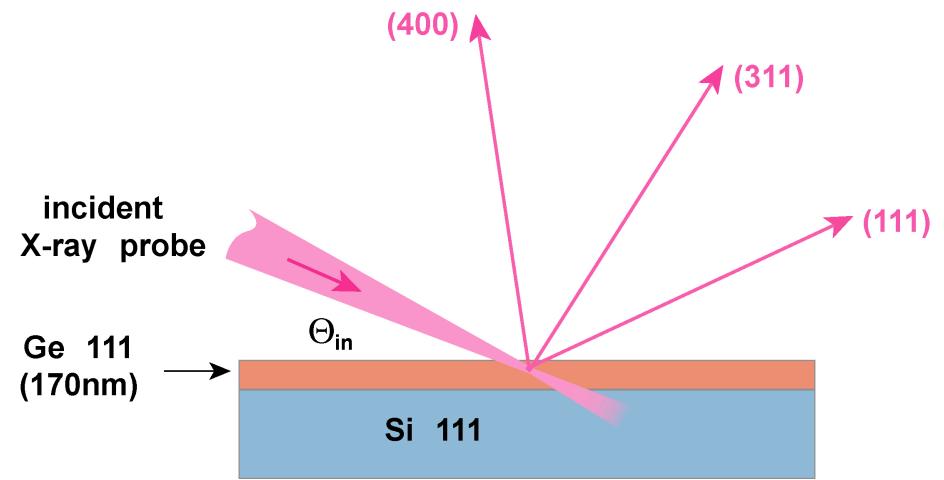
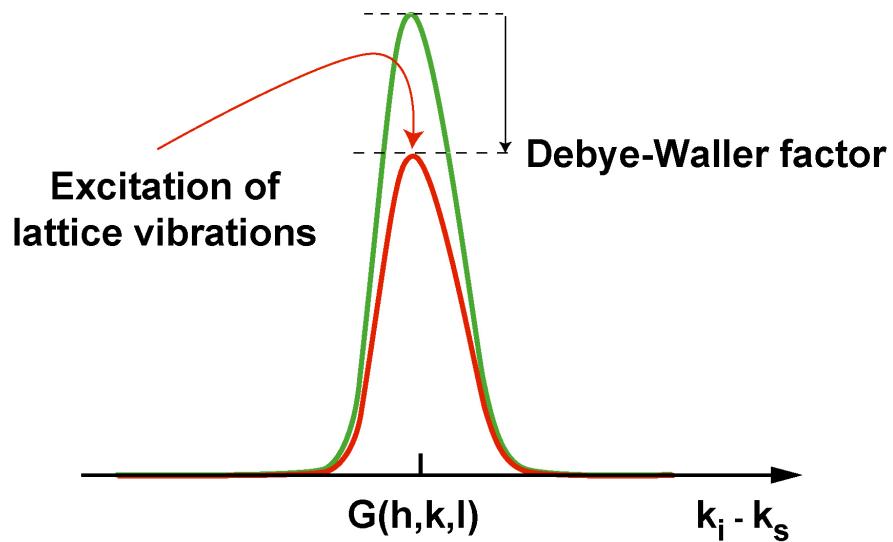


electr. excitation removes **barriers**

non-thermal melting is a  
**2nd-order** like phase transition,  
but what's the pathway ??

It is necessary to measure **atomic displacements** !

# Measuring Atomic Displacements (I): *Transient Debye-Waller Effect*



$$DWF = \exp\left(-\frac{4}{3}\pi^2 \langle u^2 \rangle / d_{hkl}^2\right)$$

*mean atomic displacement*

111-surface:

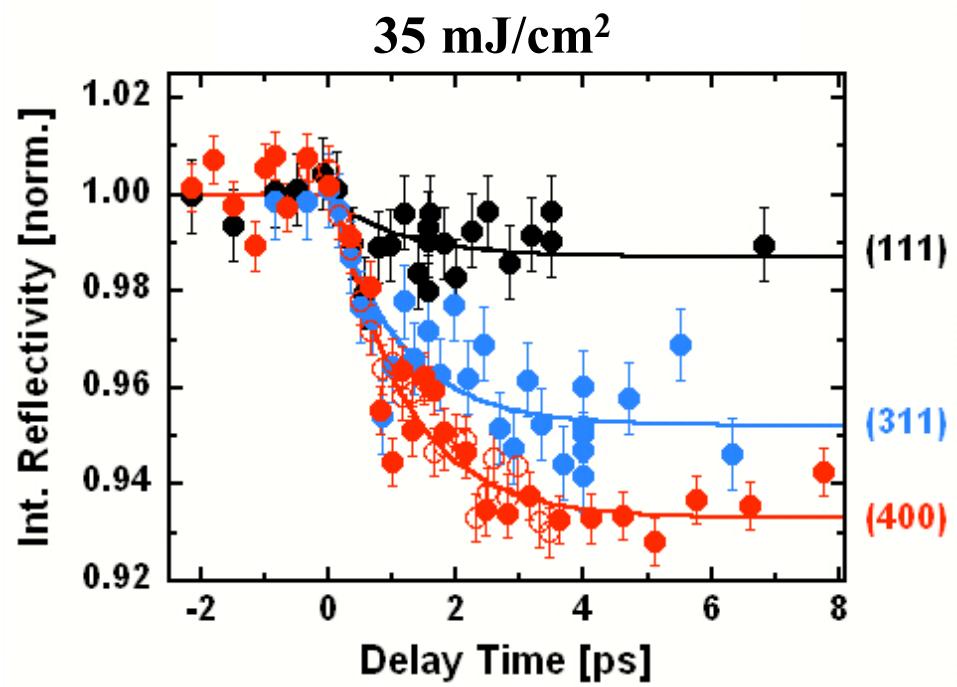
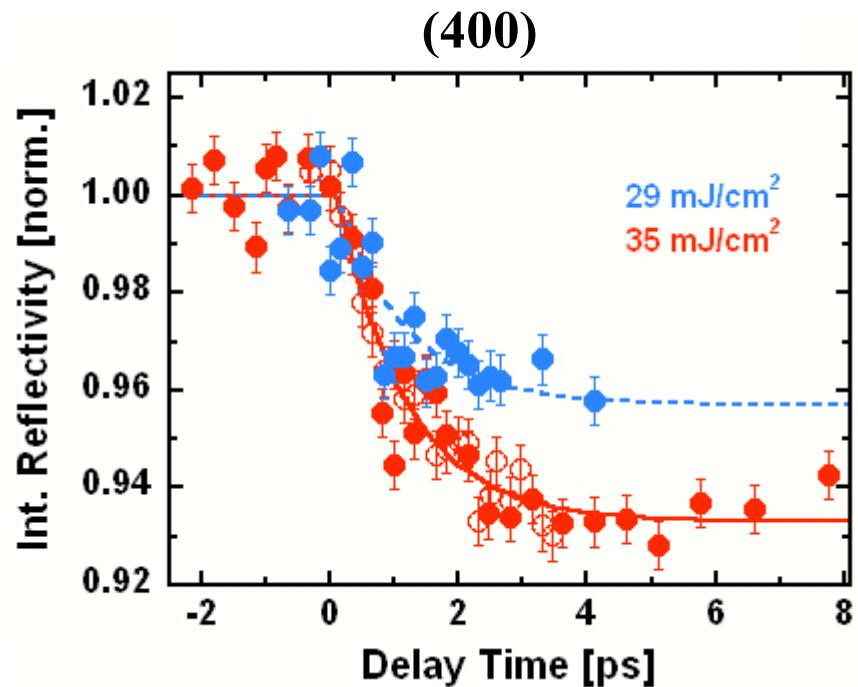
111-refl.:  $\Rightarrow \Theta_{in} = 24.9^\circ; \Theta_{out} = 24.9^\circ$

311-refl.:  $\Rightarrow \Theta_{in} = 24.2^\circ; \Theta_{out} = 48.8^\circ$

400-refl.:  $\Rightarrow \Theta_{in} = 21.6^\circ; \Theta_{out} = 96.8^\circ$

# Electron-to-Lattice Energy Relaxation

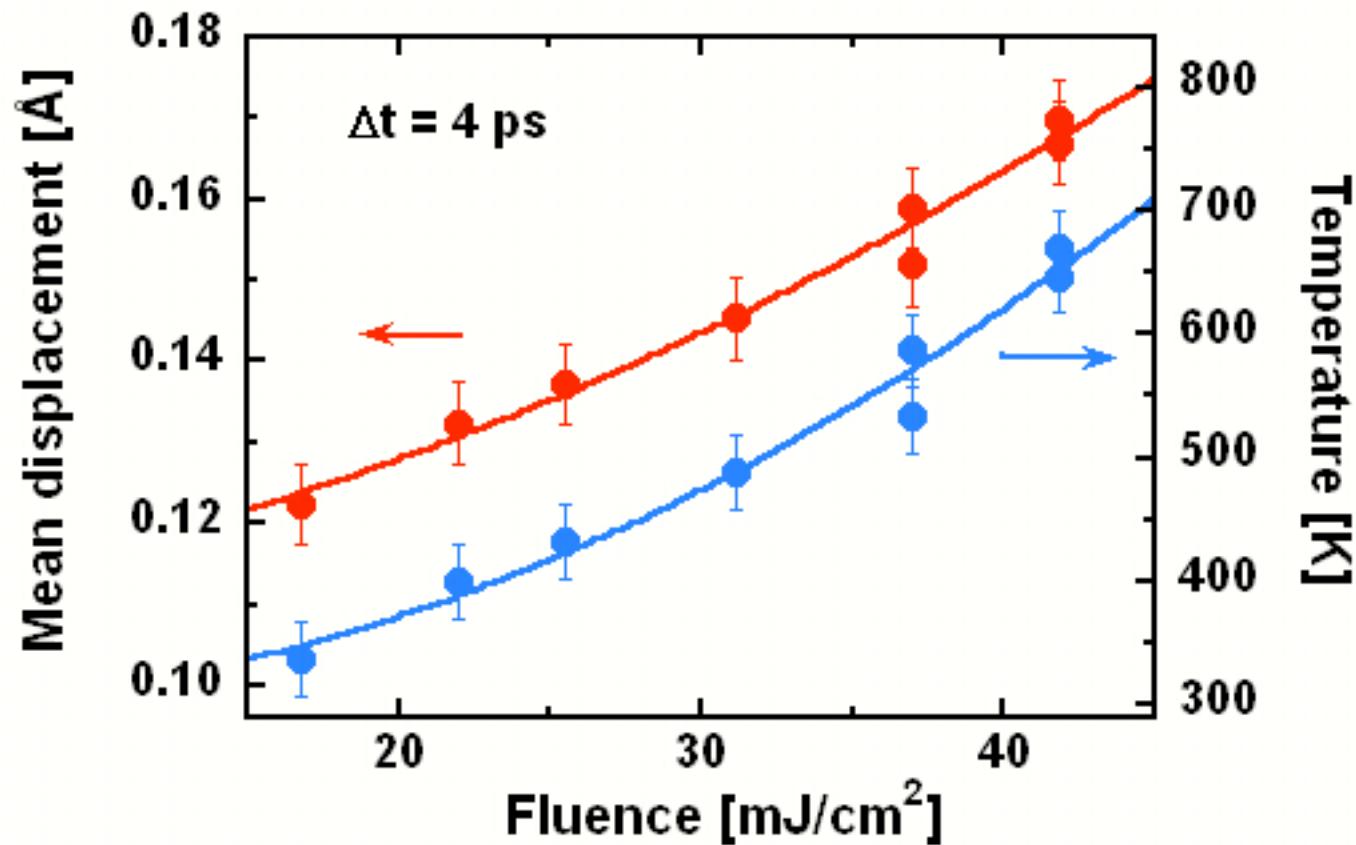
170nm Ge (111) on Si (111)



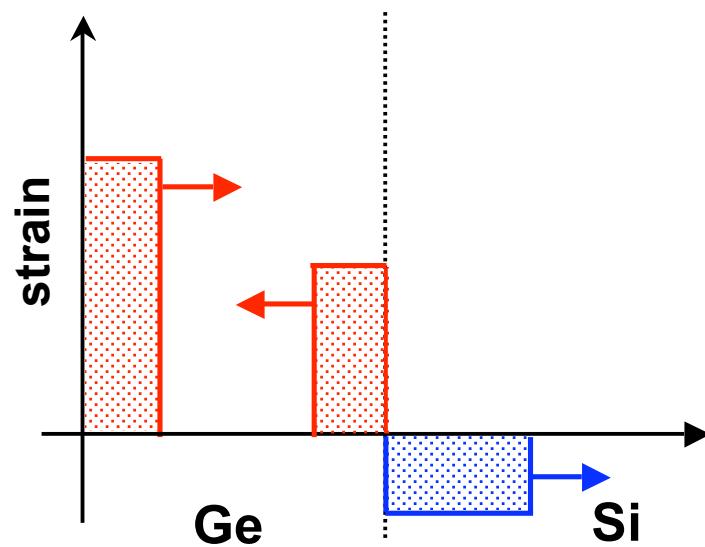
*energy relaxation time: 1.1 ps*

*it is Debey-Waller !!*

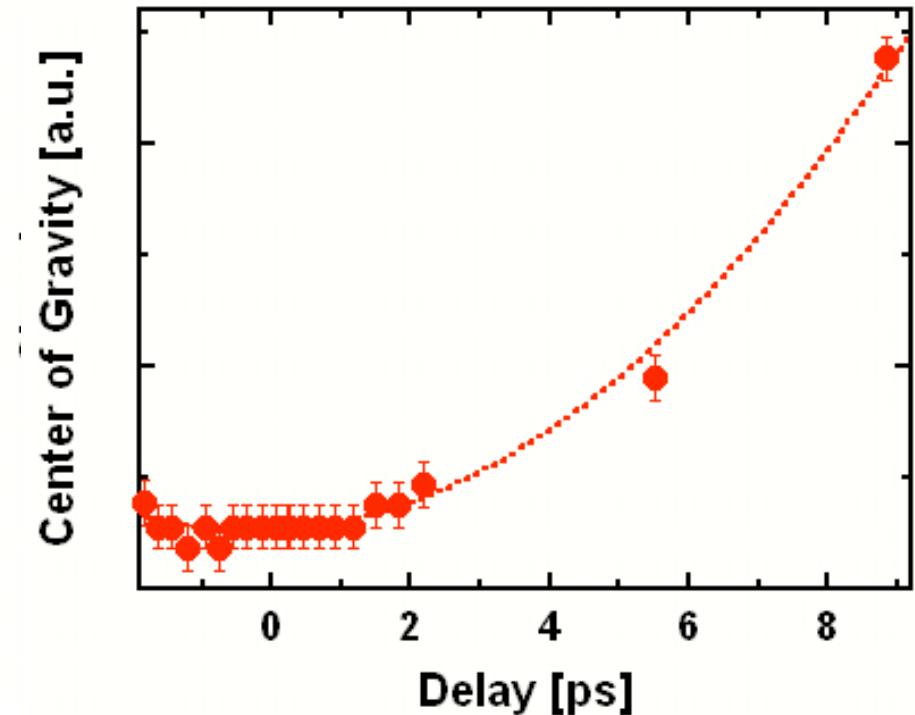
# Mean Displacement $\leftrightarrow$ Temperature



# Thermoacoustic Response: *Coherent Acoustic Phonons*



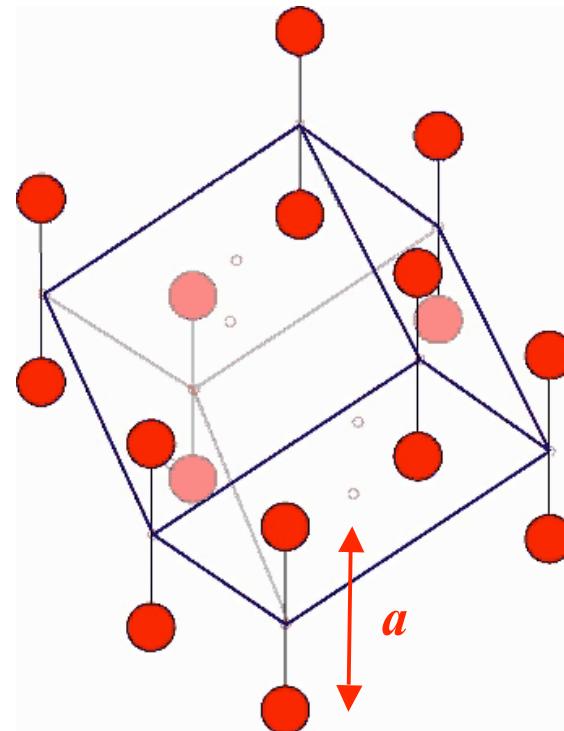
A. Cavalleri et al., PRL 85, 586 (2000)



No *electronic stress contribution* !

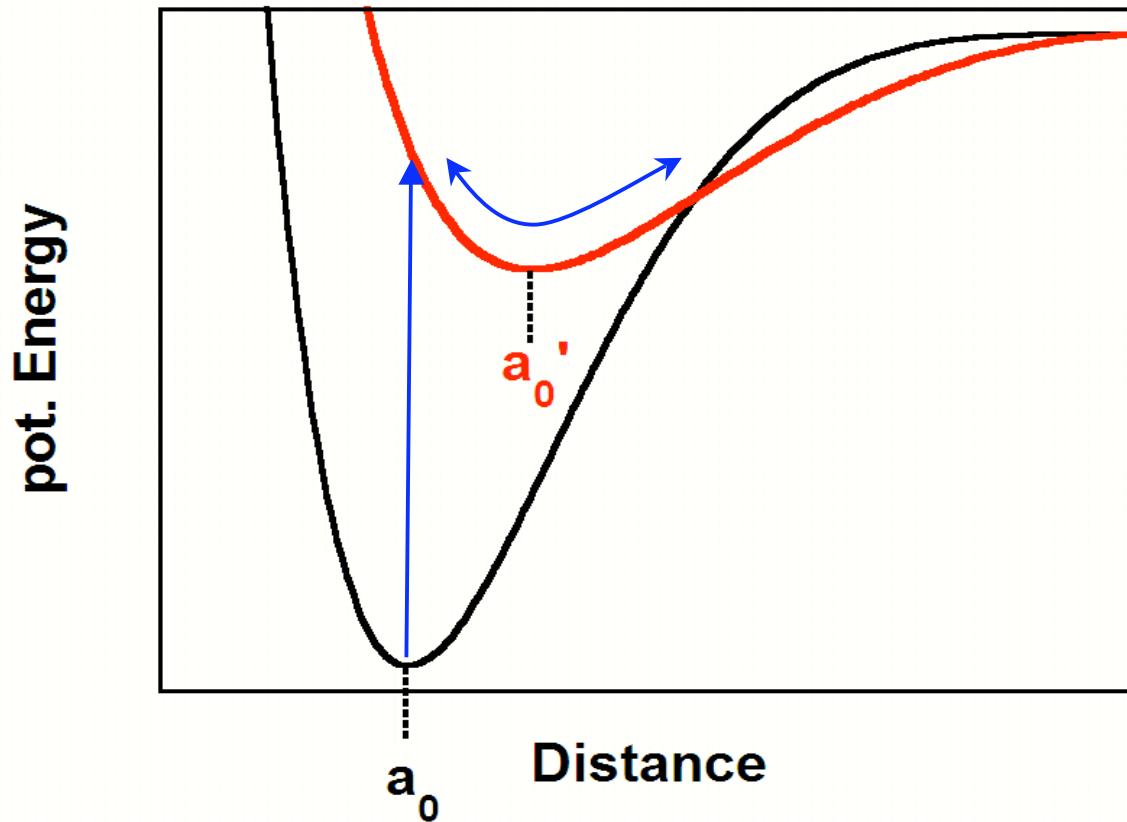
# Measuring Atomic Displacements (II): *Coherent Optical Phonons in Bismuth*

- semi-metal
- rhombohedral structure:
  - slightly distorted fcc
  - di-atomic basis
- excitation of *coherent optical phonons* ( $A_{1g}$ -mode) with fs-pulses
  - Zeiger et al., PRB **45**, 768 (1992)
  - DeCamp et al., PRB **64**, 92301 (2001)
  - Hase et al., PRL **88**, 67401 (2002)

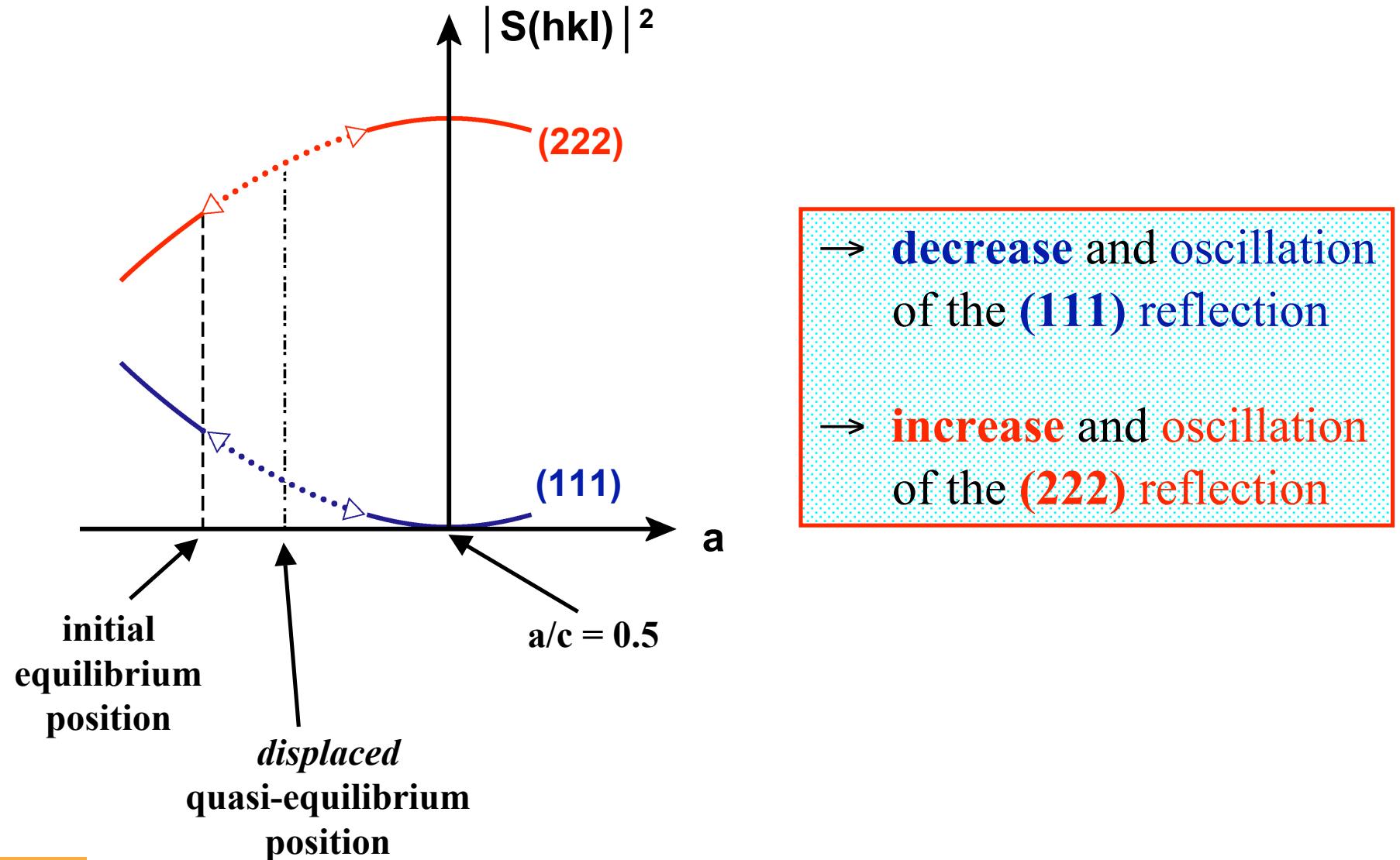


Important:  
*Peierls-Jones distortion*  
↔  
 $A_{1g}$ -phonon

# Displacive Excitation of Coherent Phonons

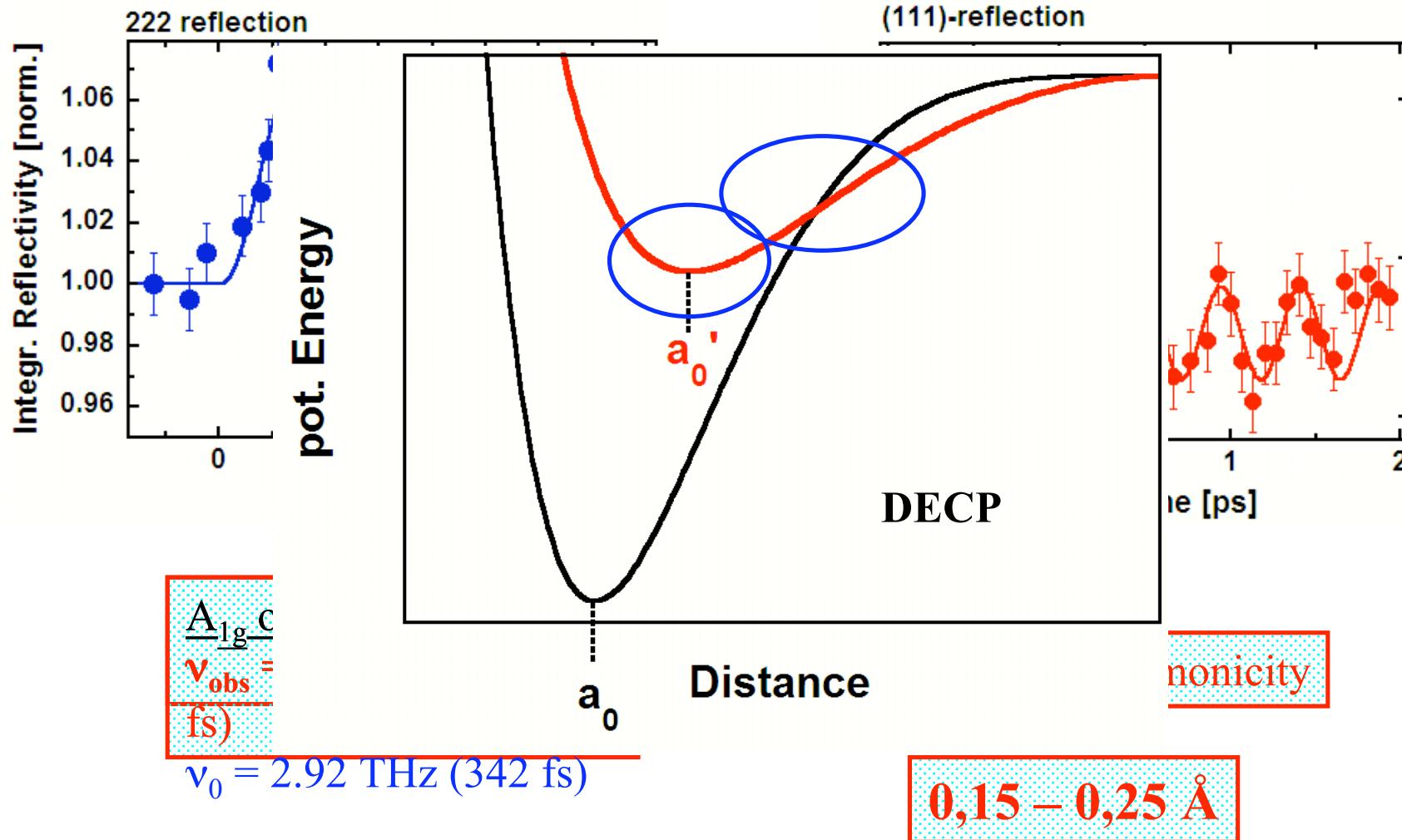


# Geometrical Structure Factor of Bismuth



# Coherent Optical Phonons

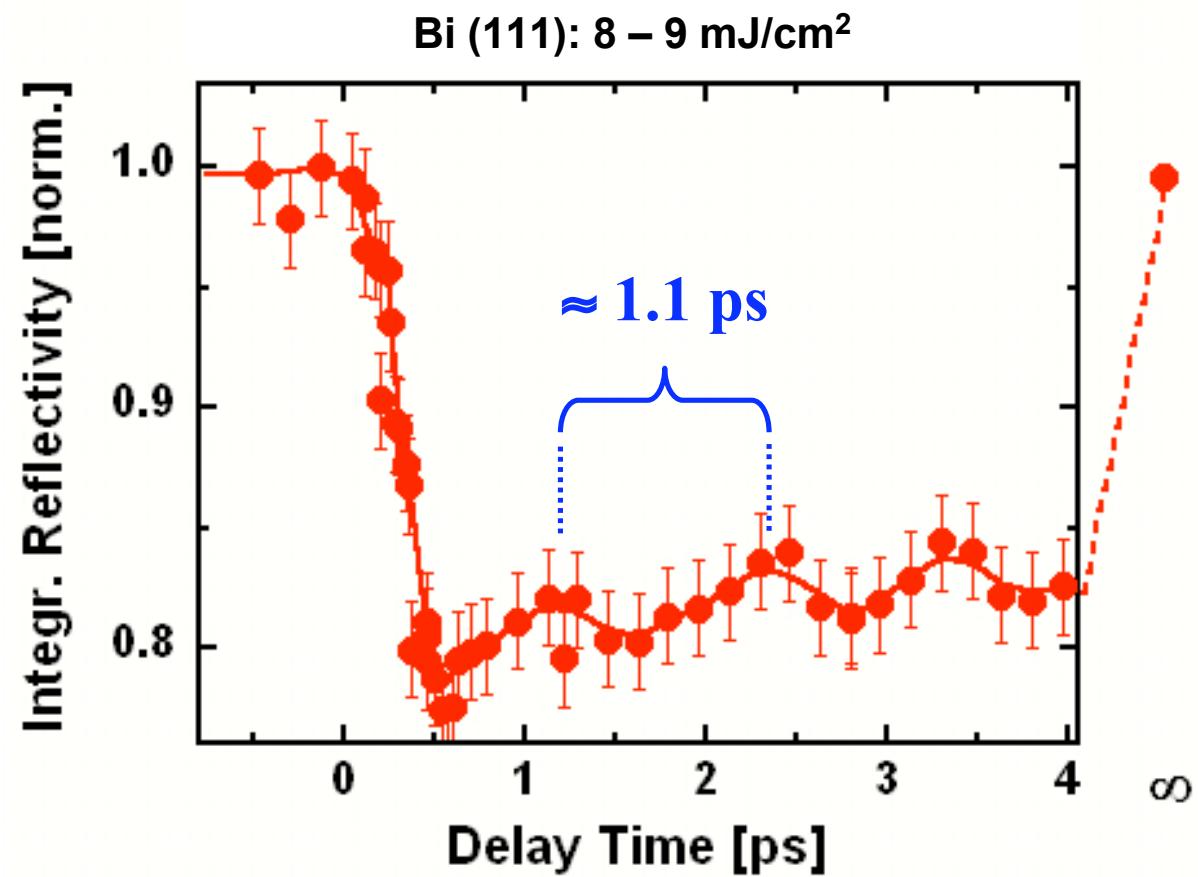
Bi 50nm on Si,  $F \approx 6 \text{ mJ/cm}^2$



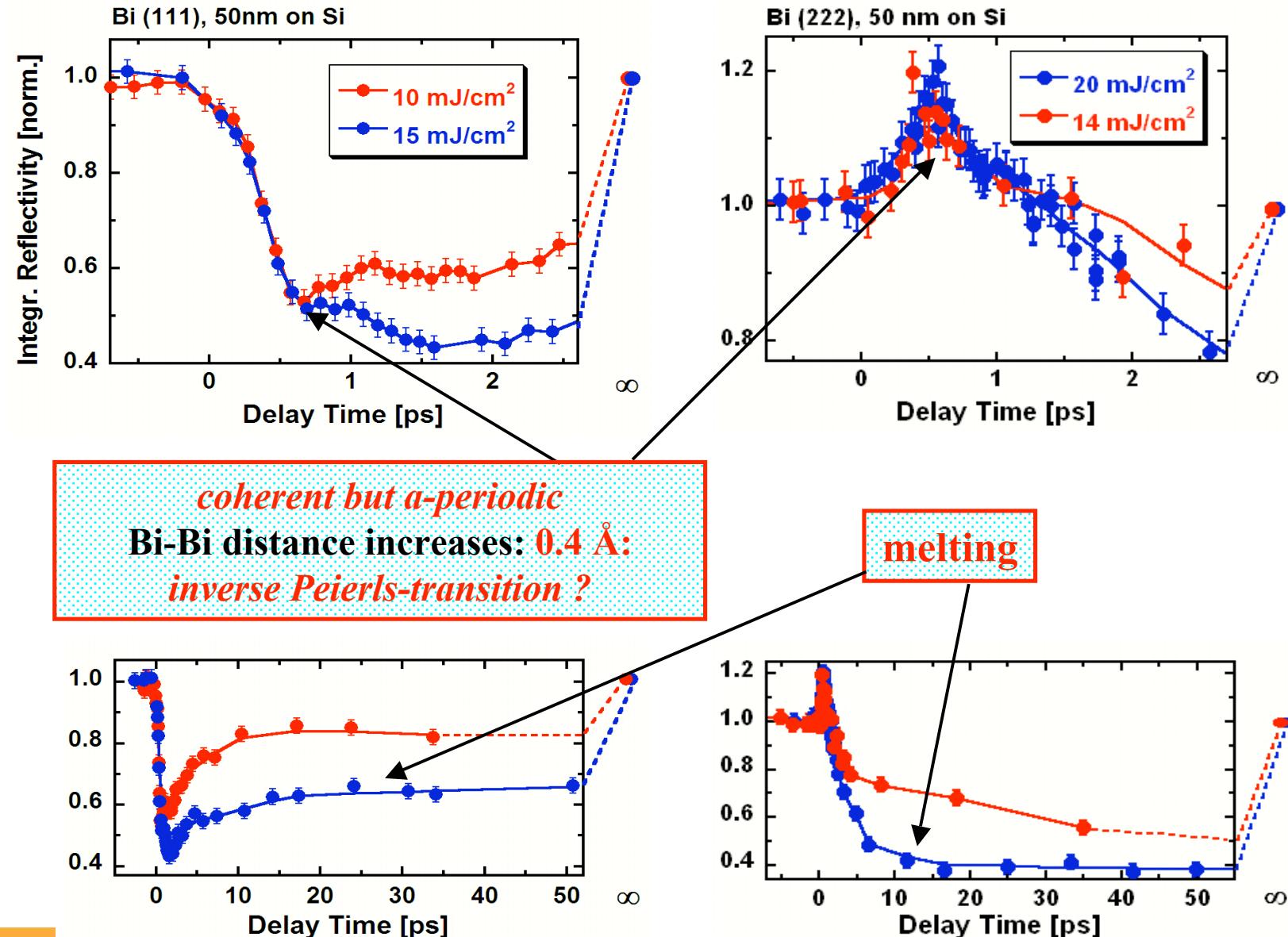
S. Fahy, poster #17

K. Sokolowski-Tinten et al., Nature 422, 287 (2003)

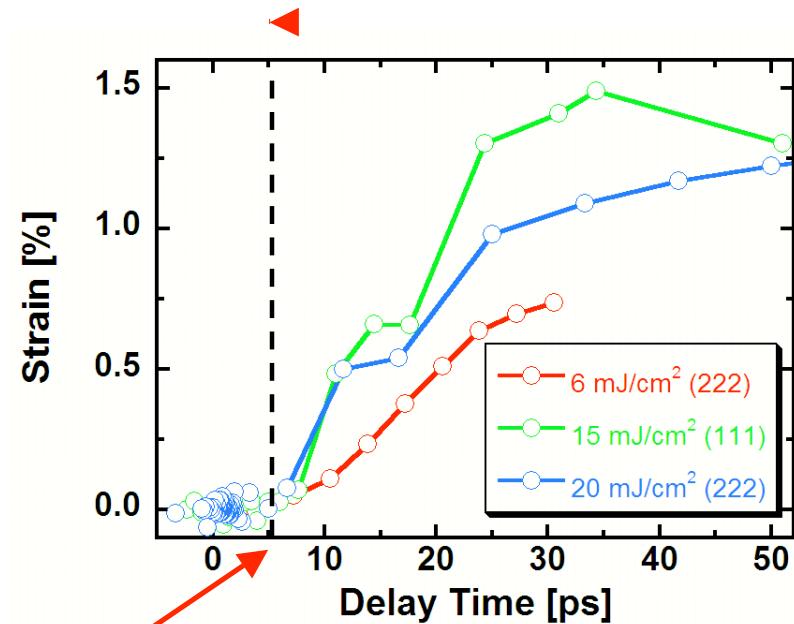
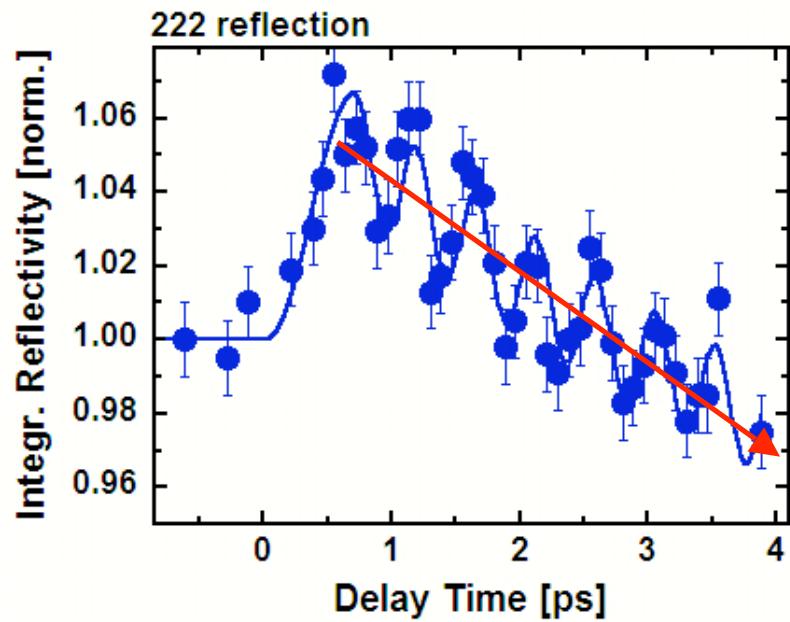
# Phonon Softening



# High Fluences: *Phase Transition(s)* in Bismuth



# Energy Relaxation in Bismuth



delayed thermal expansion

slower than in Ge; again no electronic stress.

# Acknowledgements

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# Thank you!